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Green's function theory of cubic antiferromagnet with anisotropic exchange

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Some of the statistical mechanical properties of two-sublattice cubic antiferromagnets with anisotropic exchange have been discussed. After transforming the usual spin-Hamiltonian to a Bose operator Hamiltonian by Dyson-Malcev transformation, the Green's function methods have been applied with properly chosen decoupling scheme to derive the magnon energy spectrum, the sublattice magnetization and the relation between the Neel temperature and the anisotropy parameter η. Results for $S = 1$, $3/2$, $5/2$, $7/2$ have been studied and it is shown that the general nature of variation of T with η resembles closely with the variation of Curie temperature with the antisotropy parameter for ferromagnets. The heat capacity for antiferromagnetic magnons has been derived and is found to obey a $\eta^{-3/2}$ law, while the spinwave contribution to thermal conductivity is roughly proportional to $1/\eta$.

1. INTRODUCTION

The role played by the anisotropic exchange in antiferromagnets has not been discussed in much detail in the literature and the main stream of researches was confined to the study of the statistical mechanical properties of those antiferromagnets whose spins are coupled by the isotropic nearest neighbour Heisenberg exchange interaction (Li 1951, Pu 1960, Anderson & Callen 1964, Honma 1963, Lines 1963, 1964, 1965, Lines & Jones 1966, Lee & Liu 1967, Oguchi 1973). From both theoretical and experimental points of view it is, however, important to investigate the effect of the anisotropic exchange on statistical mechanics of antiferromagnets. The most advantageous way of studying such effect is to use the double time temperature dependent Green's function formalism which has been applied with some success to the statistical problems in magnetism. The chief motivation of the present paper is to derive, within the Green's function framework, the magnon energy spectrum, the sublattice magnetization, expressions for spin wave thermal conductivity and the magnon heat capacity and to find out the variation of the so-called Neel temperature with the anisotropic exchange. The entire investigation is based on a model which considers a two-sublattice antiferromagnetic system, consisting of a simple cubic or a body centered or a face-centered cubic array of magnetic ions of spin S interacting via an anisotropic exchange interaction. The procedure involves the transformation of the usual spin Hamiltonian into a Bose operator Hamiltonian and the use of double time temperature dependent Green's functions. The main difficulty in using the Green's functions is that the Green's function equation of motion involves some higher order Green functions. The problem related to the linearization of the equation of motion has been resolved by means of adequate decoupling approximations. After solving the equation of motion various thermodynamic properties have been discussed. The variation of Neel temperature for $S = 1$, $3/2$, $5/2$, and $7/2$ has been graphically studied restricting to the cubic lattices. The general nature of variation of Neel temperature T_N with the anisotropy parameter η is found to be similar to the variation of Curie temperature with the anisotropy parameter for ferromagnetic lattices as shown by Flax & Raich (1969). In addition to this aspect of variation of T_N with η, expressions for magnon heat capacity C_m and the magnon thermal conductivity K_s have been derived and it has been observed that both these physical quantities obey some approximate laws : $C_m \propto \eta^{-3/2}$ and $K_s \propto \eta^{-1}$.

2. Dyson-Maleev Transformation

The Hamiltonian for a two sublattice antiferromagnet with nearest neighbour anisotropic exchange may be written in the following form

$$H = 2J \sum_{<ij>} [S_{1j}^z S_{2i}^z + \tfrac{1}{2}\eta(S_{2i}^+ S^-_{1j} + S^-_{2i} S_{1j}^+)], \tag{1}$$

where η is the anisotropy parameter whose value is assumed to lie between 0 and 1. The symbol $1j(2i)$ refers to the lattice site $j(i)$ in the sublattice 1(2) and the summation $\Sigma_{<ij>}$ runs over all nearest neighbour pairs. J is the nearest-neighbour exchange constant. It is also assumed that the magnetization in the sublattice 1 is in the $+z$ direction and that in the sublattice 2 is in the $-z$ direction.

The spin operators satisfy the usual commutation relations

$$(S_l^\pm, S_{l'}^z] = \mp S_l^\pm \delta_{ll'}; \quad [S_l^-, S_{l'}^+] = -2S_l^z \delta_{ll'} \quad \dots \tag{2}$$

where l, l' stand for $1j$ or $2i$.

We now transform the spin-Hamiltonian expressed by eq. (1) into a Bose operator Hamiltonian. The operators we introduce are b_{1j}, b_{1j}^+, a_{2i}, a_{2i}^+ which satisfy the following commutation relations

$$\begin{aligned}
&[(a_{2i}(b_{1j}), a_{2i'}^+(b_{1j'}^+)] = \delta_{2i(1j);\, 2i'\,(1j')},\\
&[a_{2i}(b_{1j}), a_{2i'}(b_{1ij'})] = [a_{2i}^+(b_{1j}^+), a_{2i'}^+(b_{1j'}^{\ +})] = 0, \qquad (2a)\\
&[a_{2i}, b_{1j}(b_{1j}^+)] = [a_{2i}^+, b_{1j}(b_{1j}^+)] = 0.
\end{aligned}$$

The physical significance of the boson operators employed above is that a_{2i}^+ and a_{2i} respectively create and destroy bosons at the lattice site i in the sublattice 2 and b_{1j}^+, b_{1j} respectively create and destroy bosons at the lattice site j in the sublattice 1. In contrast with this, the spin operators by which the fundamental Hamiltonian has been expressed have no such simple physical meaning. In fact, the spin waves in a ferromagnet are conceived in terms of propagation of so-called *spin deviation*. Dyson (1956) and Maleev (1957) translated this concept of spin deviation into Bose operator language. The Dyson-Maleev transformation as applied to antiferromagnetic spin system may be written in the form

$$S_{1j}^+ = \epsilon b_{1j}, \qquad S_{2i}^+ = \epsilon a_{2i}^+,$$

$$S_{1j}^- = \epsilon b_{1j}^+\left(1-\frac{1}{\epsilon^2}b_{1j}^+b_{1j}\right), \qquad S_{2i}^- = \epsilon\left(1-\frac{1}{\epsilon^2}a_{2i}^+a_{2i}\right)a_{2i}, \qquad \dots \quad (3)$$

$$S^z{}_{1j} = \frac{\epsilon^2}{2}-b_{1j}^+b_{1j}, \qquad S^z{}_{2i} = -\frac{\epsilon^2}{2}+a_{2i}^+a_{2i},$$

where

$$\epsilon = (2S)^{\frac{1}{2}}.$$

Utilizing the transformation (3), eq. (1) therefore takes the following form

$$H = J\epsilon^2 \sum_{<ij>}\left(a_{2i}^+a_{2i}+b_{1j}^+b_{1j}+\eta a_{2i}b_{1j}+\eta a_{2i}^+b_{1j}^+-\frac{\epsilon^2}{2}\right)$$

$$-J\sum_{<ij>}(2a_{2i}^+a_{2i}b_{1j}^+b_{1j}+\eta b_{1j}a_{2i}^+a_{2i}a_{2i}+\eta b_{1j}^+b_{1j}^+b_{1j}a_{2i}^+). \qquad \dots \quad (4)$$

The Bose operator Hamiltonian written above will be employed to derive the Green's function equation of motion in the following section.

3. Green's Function Equations

In a varied selection of problems in magnetism the application of double time temperature dependent Green's function has yielded many fruitful results (Lines 1963, 1964, 1965; Tahir-Kheli 1967; Callen 1963; Lovesey 1968; Oguchi & Honma 1969; Oguchi 1963, 1973; Nagai & Tanaka 1969; Lee & Liu 1967; Watarai & Kawasaki 1972; Woolsey & White 1969; Chakraborty 1974, 1975). These Green's function involving two Bose operators A and B may be defined as

$$G_{AB}(t) = \langle\langle A; B\rangle\rangle = -i <TA(t)B(0)>,$$

where the angular brackets denote ensemble averages and T the Dyson time ordering operator

$$TA(t)B(0) = \theta(t)A(t)B(0)+\theta(-t)B(0)A(t),$$

where $\theta(x)$ is the step function defined by

$$\begin{aligned}\theta(x) &= 1 \text{ for } x > 0,\\ &= 0 \text{ for } x < 0.\end{aligned}$$

In the above definition of the Green's function we have adopted units for which the Planck's constant h is equal to 2π.

The equation of motion for the Green's function is

$$i\left(\frac{d}{dt}\right)G_{AB}(t) = \delta(t)\langle[A, B]\rangle - i\langle\langle[A(t), H(t)]B(0)\rangle\rangle. \tag{5}$$

The final term in the above equation consists, in general, of some higher order Green's functions which have to be decoupled for solving the equation of motion.

The definition of the real time Green's function as given above may be extended to the imaginary time domain by analytic continuation which is equivalent to replacing t by $-i\tau$ and $G_{AB}(t)$ by $-iG_{AB}(\tau)$. In the present problem the Green's functions of interest are (Watarai & Kawasaki 1972)

$$\begin{aligned}
G_{1j,\,1j'}(\tau) &= \langle\langle b_{1j}\,;\, b_{1j'}{}^{+}\rangle\rangle = \langle Tb_{1j}(\tau)b_{1j'}{}^{+}(0)\rangle,\\
G_{1j,\,2i}(\tau) &= \langle\langle b_{1j}\,;\, a_{2i}\rangle\rangle = \langle Tb_{1j}(\tau)a_{2i}(0)\rangle,\\
G_{2j,\,1j}(\tau) &= \langle\langle a_{2i}{}^{+}\,;\, b_{1j}{}^{+}\rangle\rangle = \langle Ta_{2i}{}^{+}(\tau)b_{1j}(0)\rangle,\\
G_{2i,\,2i'}(\tau) &= \langle\langle a_{2i}{}^{+}\,;\, a_{2i'}{}^{+}\rangle\rangle = \langle Ta_{2i}{}^{+}(\tau)a_{2i'}{}^{+}(0)\rangle,
\end{aligned} \tag{6}$$

defined in terms of imaginary time argument τ. Using the boson commutation relations and eq. (4), the equation of motion for $G_{1j,\,1i'}(\tau)$ may be written as

$$\begin{aligned}\left(\frac{d}{d\tau}\right) G_{1j,\,1j'}(\tau) = \delta(\tau)\delta_{jj'} - J\epsilon^2 \sum_{\langle ij\rangle} [\langle\langle b_{1j}\,;\, b_{1j'}{}^{+}\rangle\rangle + \eta\langle\langle a_{2i}{}^{+}\,;\, b_{1j'}{}^{+}\rangle\rangle]\\
+2J\sum_{\langle ij\rangle} [\langle\langle a_{2i}{}^{+}a_{2i}b_{1j}\,;\, b_{1j'}{}^{+}\rangle\rangle + \eta\langle\langle b_{1j}{}^{+}a_{2i}{}^{+}b_{1j}\,;\, b_{1j'}{}^{+}\rangle\rangle. \quad \ldots\end{aligned} \tag{7}$$

The final term in the above equation consists of complicated four boson Green's functions. The usual procedure to solve eq. (7) is to linearize it by decoupling these four-boson Green's functions to two-boson Green's function. There exists in the literature a large number of papers dealing with this decoupling problem. Usually some heuristic approximations are made to decouple the higher order Green's functions. Unfortunately, one has yet no exact knowledge of the error involved in any decoupling approximation and hence for the assessment of the quantitative validity of the approximation one has to compare the final results with the available experimental data. In some cases, however, suitable experimental information is lacking and in those cases the decoupling approximations should be devised on the basis of some concrete logic. This situation has occurred in the study of the biquadratic exchange in magnetic systems and has been tackled by the author (Chakraborty 1975) by some mathematically justified decoupling schemes. The Green's functions occurring in eq. (7) are of familiar form. These Green's functions can be decoupled by the following heuristic approximations :

$$\langle\langle a_{2i}^{+}a_{2i}b_{1j}\,;\,b_{1j'}^{+}\rangle\rangle \rightarrow \langle a_{2i}^{+}a_{2i}\rangle\langle\langle b_{1j}\,;\,b_{1j'}^{+}\rangle\rangle + \alpha_{1j}\langle a_{2i}b_{1j}\rangle\langle\langle a_{2i}^{+}\,;\,b_{1j'}^{+}\rangle\rangle, \quad \ldots \text{(8a)}$$

$$\langle\langle b_{1j}^{+}a_{2i}^{+}b_{1j}\,;\,b_{1j'}^{+}\rangle\rangle \rightarrow \langle b_{1j}^{+}b_{1j}\rangle\langle\langle a_{2i}^{+}\,;\,b_{1j'}^{+}\rangle\rangle + \alpha_{2i}\langle b_{1j}^{+}a_{2i}^{+}\rangle\langle\langle b_{1j}\,;\,b_{1j'}^{+}\rangle\rangle \quad \ldots \text{(8b)}$$

where α_{1j} and α_{2i} may be called the Callen parameters. If one makes a choice $\alpha_{1j} = \alpha_{2i} = 1$ the decoupling approximations become identical with those of Watarai & Kawasaki (1972). Such a decoupling approximation has been called as random phase approximation. We do, however, make a more realistic choice

$$\alpha_{1j} = \frac{\langle S_{1j}^{z}\rangle}{2S^2}, \quad \alpha_{2i} = -\frac{\langle S_{2i}^{z}\rangle}{2S^2}. \quad \ldots \text{(9)}$$

As will be seen later such a choice enables one to recover the results of Lee & Liu (1967) for the special case $\eta = 1$. It is apparent that at very low temperatures $\langle S_{1j}^{z}\rangle \sim S$ and $\langle S_{2i}\rangle \sim -S$. so that for $S = \frac{1}{2}$, one has $\alpha_{1j} = \alpha_{2i} = 1$. Therefore the decoupling approximation (8a) and (8b) are the modifications over the random phase approximation.

Using eq. (8) along with choice (9) we obtain the equation of motion in the following form :

$$\left(\frac{d}{d\tau}\right) G_{1j,\,1j'}(\tau) = \delta(\tau)\delta jj' - J\epsilon^2 \sum_{j} (G_{1j,\,1j'}(\tau) + \eta G_{2i,\,1j'}(\tau))$$

$$+2J\sum_{j'}\{(\langle a_{2i}^{+}a_{2i}\rangle - \frac{\eta}{2S^2}\langle S_{2i}^{z}\rangle\,\langle b_{1j}^{+}a_{2i}^{+}\rangle)G_{1j,\,1j'}(\tau)$$

$$+(\eta\langle b_{1j}^{+}b_{1j}\rangle + \frac{1}{2S^2}\langle S_{1j}^{z}\rangle\,\langle a_{2i}b_{1j}\rangle)G_{2i,\,1j'}(\tau)\}. \quad \ldots \text{(10)}$$

For $\eta = 1$ this equation reduces to that of Lee & Liu (1967).

Since the lattice is translationally invariant, one can use the Fourier transforms

$$G_{1j,\,1j'}(\tau) = \frac{2}{N\beta}\sum_{k,l} G_{11}(\boldsymbol{k}, l)\exp\left[i\boldsymbol{K}(\boldsymbol{R}_j - \boldsymbol{R}_{j'}) - i\omega_l\tau\right], \quad \ldots \text{(11)}$$

$$\langle a_{2i}b_{1j}\rangle = \frac{2}{N}\sum_{k}\langle a_{2,k}b_{1,-k}\rangle\exp\left[-i\boldsymbol{k}(\boldsymbol{R}_i - \boldsymbol{R}_j)\right], \quad \ldots \text{(12)}$$

where

$$\omega_l = 2\pi l\beta^{-1} \quad \ldots \text{(13)}$$

l being an integer, the sum over $\boldsymbol{k}$ runs over N points of the first zone in k space. $\beta = 1/k_BT$, k_B being the Boltzman constant. The equation of motion can then be transformed into

$$-i\omega_l G_{11}(\boldsymbol{k}, l) = 2m_1 + \lambda_2(0, \eta)G_{11}(\boldsymbol{k}, l) - \eta^{-1}\lambda_1(\boldsymbol{k}, \eta)G_{21}(\boldsymbol{k}, l) \\ -(\eta-\eta^{-1})\lambda_1(\boldsymbol{k}, 0)G_{21}(\boldsymbol{k}, l). \quad \ldots \ (14)$$

Proceeding in the same manner one can obtain the equations of motion G_{11}, G_{22}, G_{12} and G_{21}, G_{22}, G_{12}. In general,

$$-i\omega_l G_{AB}(\boldsymbol{k}, l) = 2m_A\delta_{AB} + \lambda_C(0, \eta)G_{AB}(\boldsymbol{k}, l) - \lambda_A(\boldsymbol{k}, \eta)G_{CB}(\boldsymbol{k}, l) \\ -(\eta-\eta^{-1})\lambda_A(\boldsymbol{k}, 0)G_{CB}(\boldsymbol{k}, l); \quad A \neq C, \quad \ldots \ (15)$$

where m_A stands for $<S_{Al}^z>$ and

$$\lambda_A(\boldsymbol{k}, \eta) = 2m_A J(\boldsymbol{k})\left[1 - \frac{\eta}{2S^2}\sum_{\boldsymbol{k}'}\gamma(\boldsymbol{k}')<a_2\boldsymbol{k}'b_1, {}_{-}\boldsymbol{k}'>\right], \quad \ldots \ (16)$$

$$J(\boldsymbol{k}) = J\sum_{\delta}\exp(i\boldsymbol{k}.\delta), \quad \ldots \ (17)$$

$$\gamma(k) = \frac{1}{z}\sum_{\delta}\exp(i\boldsymbol{k}.\delta), \quad \ldots \ (18)$$

z being the number of nearest neighbours and δ a nearest neighbour vector.

4. Magnon Dispersion Relation

Due to the structural equivalence of two sublattices, one obtains the symmetric relation

$$\lambda_1(\boldsymbol{k}, \eta) = -\lambda_2(\boldsymbol{k}, \eta) = \gamma(\boldsymbol{k})\lambda(0, \eta). \quad \ldots \ (16a)$$

The solution of eq. (15) may be obtained in straight forward manner. One finds

$$G_{11}(\boldsymbol{k}, l) = 2m_1\frac{i\omega_l + \lambda(0, \eta)}{\omega_{\boldsymbol{k}, \eta}{}^2 + \omega_l{}^2}, \quad \ldots \ (19)$$

$$G_{22}(\boldsymbol{k}, l) = 2m_2\frac{i\omega_l - \lambda(0, \eta)}{\omega_{\boldsymbol{k}, \eta}{}^2 + \omega_l{}^2}, \quad \ldots \ (20)$$

$$G_{12}(\boldsymbol{k}, l) = 2m_2\frac{\lambda(\boldsymbol{k}, \eta)}{\omega_{\boldsymbol{k}, \eta}{}^2 + \omega_l{}^2}, \quad \ldots \ (21)$$

$$G_{21}(\boldsymbol{k}, l) = -2m_1\frac{\lambda(k, \eta)}{\omega_{\boldsymbol{k}, \eta}{}^2 + \omega_l{}^2}, \quad \ldots \ (22)$$

where $\omega_{\boldsymbol{k}, \eta}$ determines the magnon energy spectrum given by

$$\omega_{\boldsymbol{k}, \eta} = \lambda(0, \eta)\left[1 - \gamma^2(k)\left\{\frac{1}{\eta} + \left(\eta - \frac{1}{\eta}\right)\frac{\lambda(0, 0)}{\lambda(0, \eta)}\right\}\right]^{\frac{1}{2}}. \quad \ldots \ (23)$$

For $\eta = 1$, above equation reduces to one determining the spinwave spectrum for an isotropic antiferromagnet as worked out by Lee & Liu (1967). Furthermore, at very low temperatures, since the interspin correlation is small, the difference $\lambda(0, 0)-\lambda(0, \eta)$ is very small so thtat we see from eq. (23) that a highly anisotropic antiferromagnet (corresponding to very small η) has a large spin wave energy. This is similar to the case of a ferromagnet, as worked out by Flax & Raich (1969). Furthermore, it is also to be observed that $\omega_{k,\eta}$ vanishes when the order parameter $m = m_1 = -m_2$ goes to zero indicating the onset of the paramagnetic phase.

5. Sublattice Magnetization and Neel Temperature

The sublattice magnetization m_A as deduced by Callen may be written in the form

$$m_A = \frac{(S-\phi_{AA})(1+\phi_{AA})^{2s+1}+(S+1+\phi_{AA})\phi_{AA}{}^{2s+1}}{(1+\phi_{AA})^{2s+1}-\phi_{AA}{}^{2s+1}}, \qquad \ldots \quad (24)$$

where

$$\phi_{AA} = \frac{2}{N}\Sigma\ \phi_{AA}(\boldsymbol{k}). \qquad \ldots \quad (25)$$

The functions $\phi_{AA}(k)$ derived in the fashion of Lee & Liu (1967) are given by

$$\phi_{11}(\boldsymbol{k}) = \tfrac{1}{2}[-1+\lambda(0, \eta)\omega_{k,\eta}{}^{-1} \coth \tfrac{1}{2}\beta\omega_{k,\eta}], \qquad \ldots \quad (26)$$

$$\phi_{22}(\boldsymbol{k}) = \tfrac{1}{2}[-1-\lambda(0, \eta)\omega_{k,\eta}{}^{-1} \coth \tfrac{1}{2}\beta\omega_{k,\eta}], \qquad \ldots \quad (27)$$

$$\phi_{21}(\boldsymbol{k}) = -\phi_{12}(\boldsymbol{k}) = \tfrac{1}{2}\lambda(\boldsymbol{k}, \eta)\omega_{k,\eta}{}^{-1} \coth \tfrac{1}{2}\beta\omega_{k,\eta}. \qquad \ldots \quad (28)$$

In order to evaluate $\omega_{k,\eta}$ we use a power series expansion

$$x \coth x = 1+ \sum_{n=1}^{\infty} (-1)^{n-1} \frac{2^{2n}}{(2n)\,!} B_{2n-1}x^{2n}, \quad \text{for } x^2 < \pi^2 \qquad \ldots \quad (29)$$

where B's are the Bernoulli's numbers

$$B_0 = 1, \quad B_1 = -\tfrac{1}{2}, \quad B_2 = 1/6, \quad \text{etc.}$$

Since $|\beta\omega_{k,\eta}|$ is sufficiently small (much smaller than 2π) we can utilize the above series expansion. Furthermore, we note that $|\beta\omega_{k,\eta}| \ll 1$ and therefore the series in $\coth(\frac{1}{2}\beta\omega_{k,\eta})$ starts from a term (first term) which is much larger than the subsequent terms in the series. Also since $\beta\omega_{k,\eta}$ is roughly proportional to $1/T$, the first term of the expnansion of $\coth(\frac{1}{2}\beta\omega_{k,\eta})$ becomes increasingly greater than the subsequent terms as the temperature increases. Therefore, considering only the first term of eq. (29) one can express $\lambda(0, \eta)$ from eq. (16) in the following form :

$$\lambda(0, \eta) \simeq 2J(0)m \left[1+\frac{2\eta mk_BT}{NS^2\lambda(0, \eta)} \sum_{k} \frac{\gamma^2(k)}{1-t\gamma^2(k)}\right], \qquad \ldots \quad (30)$$

where

$$t = \frac{1}{\eta} + \left(\eta - \frac{1}{\eta} \right) \frac{\lambda(0, 0)}{\lambda(0, \eta)}. \qquad \dots (31)$$

We introduce a quantity

$$x = \lambda(0, \eta)/m$$

so that from eq. (30) one finds

$$x = 2J(0) \left[1 + \frac{\eta k_B T(f_t - 1)}{2tS^2 J(0)} \right], \qquad \dots (32)$$

where

$$f_t = \frac{2}{N} \sum_k \frac{1}{1 - t\gamma^2(k)}. \qquad \dots (33)$$

Evaluation of this sum for cubic lattices has been done in the next section. In deriving eq. (32) we have also substituted $(2/N) \sum_k 1 = 1$.

Considering the first term of expansion (29) we get from eq. (25) and eq. (27)

$$\phi_{11} = -\tfrac{1}{2} + \frac{k_B T}{\lambda(0, \eta)} f_t. \qquad \dots (34)$$

If one desires to calculate the sublattices magnetization, then the full expression given by eq. (34) has to be used. But in deriving the expression for Neel temperature the first term in the right hand side of eq. (34) may be omitted because this term finally vanishes when one takes the limit $m \to 0$.

Before deriving the Neel temperature we shall approximate the expression given by eq. (24). It is an usual method to expand eq. (24) in powers of ϕ^{-1} and to take the first term of this expansion so that the result is

$$m_A = \frac{S(S+1)}{3\phi_{AA}}. \qquad \dots (35)$$

Combining the eqs. (32), (34) and (35) one gets in the limit $m \to 0$, the following expression for Neel temperature

$$\tau_N = \frac{k_B T_N}{2J(0)} = \frac{S(S+1)}{3f_\alpha} \left[1 + \frac{(S+1)\eta(f_\alpha - 1)}{3\alpha S f_\alpha} \right], \qquad \dots (36)$$

where α stands for the limit

$$\alpha = \lim_{m \to 0} (t)$$

It is to be noted that the ratio $\lambda(0, 0)/\lambda(0, \eta)$ approches unity as $m \to 0$. When one is interested in finding out the Neel temperature, this approximate limit might be accepted. This approximation provides an easy simplification of the problem, since the form of α takes a very simple form. One finds $\alpha \approx \eta$. Therefore, for $\eta = 0$ (corresponding to Ising system) the righthandside of eq. (36) reduces to $S(S+1)/3$ which is the same as the result obtained from molecular field theory. This result is exactly supported by physical reasonings. It might be observed that for $\eta = 0$, the spin correlations are dropped from the fundamental Hamiltonian leading to structure-independent collective excitations and each spin moves in an effective field which is described in the literature as a molecular field. For non-zero value of η that is when the correlation part is taken into account in the exchange Hamiltonian one obtains structure-dependent collective excitations and it will be found that for the isotropic case ($\eta = 1$) the result reduces to that of Lee & Liu (1967). Therefore, eq. (36) gives us the general qualitative result that the Neel temperature decreases with the increase of the anisotropy parameter. Aiming to justify this general qualitative result from physical standpoint we observe that if only the Ising part of the Heisenberg Hamiltonian is considered, the excitation energy of two adjacent spin deviations is lower by J than that of two non-adjacent spin deviations. This gives rise to an effective attractive interaction between spin waves leading ultimately to the formation of so-called two-magnon bound states. The fact which is interesting is that when the transverse terms in the Heisenberg Hamiltonian is included this effective attractive interaction decreases. Thus the effective interaction for $\eta = 0$ is greater than that for $\eta = 1$ which implies that with the increase of the anisotropy parameter the effective attractive interaction decreases leading to the decrease of the extent of the ordering of spin system. As a result of this, Neel temperature decreases.

6. Results for Cubic Lattices

It seems very difficult to carry out the computation of eq. (36) due to the occurrence of the quantity f_α which cannot be calculated by the conventional procedure of computing the Watson sums. The method of Flax & Raich (1969) involves a lot of algebraic difficulties. Instead of this, we follow the simple procedure as given below

We first replace the sum by an integral

$$\frac{2}{N}\sum_k \rightarrow \frac{v}{(2\pi)^3}\int d^3k,$$

where v, the volume per site, is equal to a^3, $\frac{1}{2}a^3$ and $\frac{1}{4}a^3$ for *sc*, *bcc*, and *fcc* cases respectively, a being the length of the cell edge.

$$\frac{2}{N}\sum_k \rightarrow \frac{1}{\pi^3}\int_0^\pi\int_0^\pi\int_0^\pi dxdydz,$$

where the transformed variables x, y, z are

$$x = k_x a, \quad y = k_y a, \quad z = k_z a \quad \text{for } sc,$$
$$x = \tfrac{1}{2}k_x a, \quad y = \tfrac{1}{2}k_y a, \quad z = \tfrac{1}{2}k_z a \quad \text{for } bcc,$$
$$x = \tfrac{1}{2}k_x a, \quad y = \tfrac{1}{2}k_y a, \quad z = \tfrac{1}{2}k_z a \quad \text{for } fcc.$$

Now $\gamma(k)$ for *sc*, *bcc* and *fcc* lattices are given by

$$\begin{aligned}\gamma(k) &= \tfrac{1}{3}(\cos x+\cos y+\cos z) && \text{for } sc \\ &= \cos \tfrac{1}{2}x \cos \tfrac{1}{2}y \cos \tfrac{1}{2}z && \text{for } bcc \\ &= \tfrac{1}{3}(\cos \tfrac{1}{2}x \cos \tfrac{1}{2}y+\cos \tfrac{1}{2}y \cos \tfrac{1}{2}z+\cos \tfrac{1}{2}z \cos \tfrac{1}{2}x). && \text{for } fcc.\end{aligned} \tag{37}$$

Expanding f_a as

$$f_a = \frac{2}{N} \sum_k \,[1+\eta\gamma^2(k)+\eta^2\gamma^4(k)+\dots] \tag{38}$$

and using eq. (37), we obtain the approximate series

$$\begin{aligned} f_a &= 1+0.166\eta+0.014\eta^2 && \text{for } sc, \\ f_a &= 1+0.125\eta+0.052\eta^2 && \text{for } bcc, \\ f_a &= 1+0.083\eta+0.0052\eta^2 && \text{for } fcc. \end{aligned} \tag{39}$$

Only the first three terms have been retained, other terms being neglected due to smallness.

Results computed for *sc*, *bcc* and *fcc* lattices are shown in the figures (figures 1–4) considering $S = 1, 3/2, 5/2$ and $7/2$. The general nature of variation observed

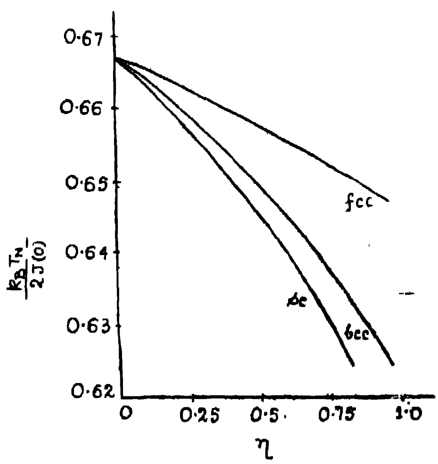

Fig. 1. The variation of Neel temperature with the anisotropy parameter for *sc*, *bcc* and *fcc* lattices for $S = 1$.

from the figures is closely analogous to the variation of ferromagnetic Curie temperature with the anisotropy parameter as established by Flax & Raich (FR). One important difference should, however, be noted. FR obtained no solution for $\eta = 0$ when they considered the Callen decoupling approximation. Of course, the aspect of indefiniteness of τ for $\eta = 0$ in the present case has been bypassed by assuming $\lambda(0,0)/\lambda(0,\eta)$ to be approximately equal to unity as m tends to zero. This assumption we have made previously and now we have obtained some justifications of the assumption. This assumption is necessary for avoiding the inconvenient singularities at $\eta = 0$.

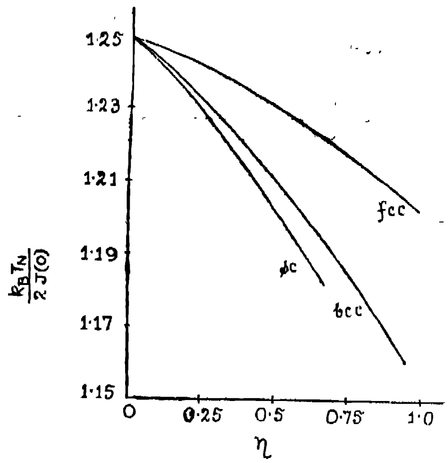

Fig. 2. The variation of Neel temperature with the anisotropy parameter for *sc*, *bcc* and *fcc* lattices for $S = 3/2$.

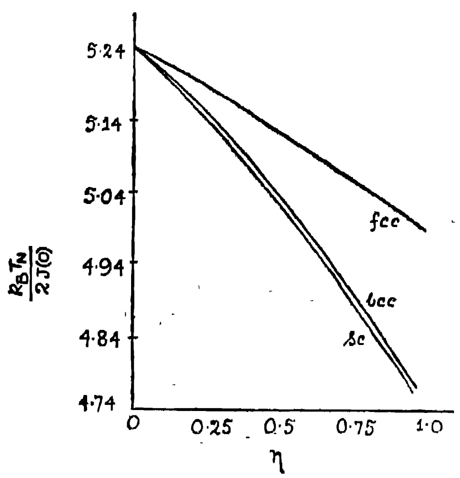

Fig. 3. The variation of Neel temperature with the anisotropy parameter for *sc*, *bcc* and *fcc* lattices for $S = 5/2$.

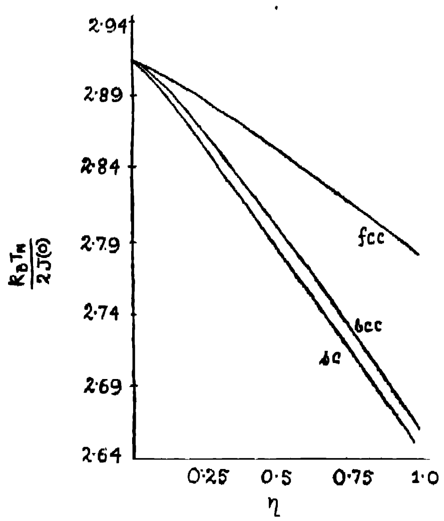

Fig. 4. The variation of Neel temperature with the anisotropy parameter for *sc*, *bcc* and *fcc* lattices for $S = 7/2$.

7. $\eta^{-3/2}$ Law for Magnon Heat Capacity

The phenomenon of heat transport by spin waves has been theoretically predicted by Frohlich & Heitler (1936) and Sato (1955) and the credit of experimental demonstration goes to Douthett & Friedberg (1961) and Douglass (1963). In fact, heat transport by spin waves is much significant at very low temperatures. Therefore, it is necessary to investigate the variation of two important thermodynamic quantities—magnon heat capacity and magnon thermal conductivity with the anisotropy paramter. These two quantities can be derived from the dispersion relation (23).

Utilizing the approximation $\lambda(0, 0) \sim \lambda(0, \eta)$ one obtains eq. (23) in the form

$$\omega_{k,\eta} = \lambda(0, \eta)[1-\eta\gamma^2(k)]^{\frac{1}{2}}. \qquad \dots (40)$$

With this spin wave spectrum we may calculate the internal energy per unit volume E

$$E = \sum_k \frac{\omega_k}{\exp(\beta\omega_k)-1}. \qquad \dots (41)$$

Replacing the sum by an integral and approximating

$$[1-\eta\gamma^2(k)]^{\frac{1}{2}} \cong 1+\tfrac{1}{2}\eta\gamma^2(k)+\dots$$

and using

$$z[1-\gamma(k)] \simeq \tfrac{1}{2}\sum_{\delta}(\boldsymbol{k}.\boldsymbol{\delta})^2 = k^2a^2,$$

one obtains after straightforward simplifications the following expression for internal energy

$$E = \left(\frac{0.601}{\beta\pi^2} - \frac{b_1}{48\beta} \right) b_2^{-3/2},$$

where

$$b_1 = \lambda(0, \eta)\left(1 - \frac{\eta}{2} \right), \quad b_2 = \frac{1}{z}\lambda(0, \eta)\beta\eta a^2,$$

other terms being neglected due to smallness. In deriving eq. (42) we have replaced m by S because the heat transport by spin waves is important at very low temperatures and we restrict ourselves to these temperatures.

Magnon heat capacity is, therefore,

$$C_m \simeq \frac{k_B^{3/2}}{2a^3\eta^{3/2}(\lambda(0, \eta))^{3/2}}\left[0.3\, k_B T^{3/2} - 3\left(1 - \frac{\eta}{2} \right) z^{3/2}\lambda(0, \eta)T^{\frac{1}{2}} \right]. \qquad \ldots (43$$

This expression shows that the specific heat for antiferromagnetic magnon, in addition to its $T^{3/2}$ dependence, has several other small temperature-dependent term, one of which is exhibited by the second term. Apart from this aspect of temperature-dependence it is to be noted that C_m obeys a $\eta^{-3/2}$ law. Thus the magnon specific heat is small for a weakly anisotropic antiferromagnet.

8. Spin Wave Thermal Conductivity

The general expression for thermal conductivity may be written in the form

$$K_S = \tfrac{1}{3} \int \frac{\partial U}{\partial T} l_s v_g dk$$

where

$$U(k, \eta, T) = \omega_{k,\eta} n^0(k, \eta, T) \frac{1}{8\pi^3} 4\pi k^2 \qquad (45)$$

is the contribution to the internal energy from spin waves of the wave vector $(k, k+dk)$, n^0 being the number of spin waves at equilibrium. The symbol l_s in eq. (44) stands for the magnon mean free path and v_g is the group velocity.

Returning to our previous expression for magnon spectrum (40) one arrives, by straight forward simplifications, at the following expression for spin wave thermal conductivity

$$K_s = (\alpha_1 k_B^3 T^2 - \alpha_2 k_B^2 T)\eta^{-1}, \qquad (46)$$

where

$$\alpha_1 = \frac{0.121z}{a^3\lambda(0, \eta)}, \quad \alpha_2 = \frac{l_s}{18a^2}\left(1 - \frac{\eta}{2} \right)$$

Eq. (46) shows that K_s is approximately proportional to $1/\eta$. This implies that the spin wave thermal conductivity is large in an anisotropic antiferromagnet. Therefore, in regard to the conductivity measurements one should choose the specimen which should possess a large anisotropy. But the difficulty lies in the fact that phonons play a significant role in an anisotropic ferromagnet and antiferromagnets and much of the heat is carried by them. This might be the possible reason of choosing the experimental specimen (for observing the heat transport by spin waves) having low anisotropy.

9. Discussion

This paper discusses some aspects of anisotropic exchange in cubic antiferromagnets on the basis of Green's function theory. In addition to the consideration of this anisotropic exchange there exists an equally important exchange interaction—the so-called biquadratic exchange interaction—which is markedly significant for some rare-earth compounds with incompletely quenched orbital angular momentum. For complete interpretation of facts, these biquadratic and anisotropic terms are to be considered along with the usual bilinear exchange Hamiltonian. The effect of biquadratic exchange has been separately discussed by the author (Chakraborty 1974, 1975). The combined effect of the anisotropic and biquadratic exchange on various statistical mechanical properties of ferromagnets and antiferromagnets has been taken up for future investigation. In all these discussions, the main mathematical apparatus is based on the elegant formalism of two-time temperature-dependent Green's functions. The art of application of Green's function theory to any specific problem is entirely dependent on the quantitative validity of the decoupling approximations, which becomes, in fact, the principal hindrance in this regard. Sufficient experimental investigations may, however, resolve this difficulty. Unfortunately, experiments conducted for finding out the effect of the biquadratic and anisotropic exchange are not sufficient. A large number of theoretical findings have been piled up during last decade and hence it is necessary to support or verify or to modify these theories by adequate experimental investigations.

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